

## R E M A R K S

Claims 11-30 are in the case.

### **Rejection under §103(a) over Goodenough et al. in view of Dallmier et al.**

Claims 11-30 stand rejected under 35 U.S.C. §103(a) as obvious over Goodenough et al. (U.S. 3,558,503) in view of Dallmier et al. (U.S. 5,683,654). Applicants respectfully request reconsideration of this rejection.

Examiner Pryor and Supervisory Patent Examiner Marschel are thanked by the undersigned for their guidance in telephonic interviews on October 26, 2010 (with Examiner Pryor), and on December 7, 2010 (with Examiner Pryor and Supervisory Patent Examiner Marschel). During the two telephonic interviews, the issues to address in the Response were clarified. The issue(s) apparently revolve around the nature and characterization of bromine chloride (BrCl).

To properly respond to the grounds of rejection in the present Office Action, the arguments from the previous Response (filed on June 23, 2010) are incorporated by reference as if set forth fully herein, to save space, and to keep the focus of the present Response on issues surrounding bromine chloride (BrCl). The remainder of the discussion below addresses the nature and characterization of bromine chloride.

In the following discussion, several auxiliary documents are referred to, and for convenience, a copy of each document (or the relevant portions thereof) accompanies this Response, even though some of these documents are already of record in this case. These documents are listed in the Table below. The left column of the Table lists these documents by Exhibit number as applied to this Response, the middle column lists the name by which each Exhibit is referred to in this Response; the right column shows the full citation for each document and, when applicable, comments.

Exhibit	Referent	Full citation and comments
1	Mills et al.	Mills et al., Bromine Chloride: an Alternative to Bromine, <i>Ind. Eng. Chem. Prod. Res. Develop.</i> , 1973, <b>12</b> , 160-165.

		Already of record; cited in a rejection in the Office Action mailed on March 4, 2009.
2	Tellinghuisen	Tellinghuisen, J. Phys. Chem. A, 2003, <b>107</b> , 753-757.
3	Second Declaration	Pages 1-4 of the Second Declaration of McKinnie, Exhibit No. 1073 in Interference 105,230. Already of record; submitted in full on Dec. 8, 2008.
4	Farkas and Lewin	Farkas and Lewin, <i>J. Am. Chem. Soc.</i> , 1950, <b>72</b> , 5766-5767.
5	Harp	Harp, D.L., <i>Current Technology of Chlorine Analysis for Water and Wastewater</i> , Hach Technical Information Series – Booklet No. 17 (2002), Page 12. Already of record; submitted on Dec. 8, 2008.
6	web page	"Halogens as Oxidizing Agents": <a href="http://www.chemguide.co.uk/inorganic/group7/halogensasoas.html">http://www.chemguide.co.uk/inorganic/group7/halogensasoas.html</a> ; accessed on December 16, 2010.
7	First Declaration	Excerpts from the Declaration of McKinnie, Exhibit No. 1001 in Interference 105,230. Already of record; submitted in full on Dec. 8, 2008.
8	Nalepa Declaration	Declaration of Christopher Nalepa Already of record; submitted on August 4, 2009.

## I. Properties of BrCl

### A. Equilibrium of BrCl

First, the nature of bromine chloride (an equilibrium mixture) and the relative amounts of the components in the equilibrium mixture are addressed. Bromine chloride is known to be in equilibrium mixture of BrCl, Br<sub>2</sub>, and Cl<sub>2</sub>, as described in Mills et al., where it is stated that "BrCl exists in equilibrium mixture containing molecular chlorine and bromine" (Page 160, last paragraph). This is reiterated in the bottom right column of Page 161, where it is stated that "Bromine chloride exists in equilibrium with bromine and chlorine in both the gas and the liquid phase", and the following equation is shown:



Mills et al. further states that "Pure bromine chloride exists only at relatively low temperatures in the solid state" (Page 161, bottom of left column). Thus, bromine chloride

is always an equilibrium mixture of BrCl, Br<sub>2</sub>, and Cl<sub>2</sub> when in the gas phase.

Tellinghuisen observes that appreciable amounts of Br<sub>2</sub> and/or Cl<sub>2</sub> are always present with BrCl for any mixing ratio of the parent gases [Br<sub>2</sub> and Cl<sub>2</sub>] (Page 753, left column, near top). The relative amounts of the three components in the equilibrium can be determined from the following equation:



At 22°C, the equilibrium constant K° is 9.1 (Page 756, left column, third paragraph of Tellinghuisen). The equilibrium expression is

$$K = [\text{BrCl}]^2 \div [\text{Br}_2][\text{Cl}_2]$$

Defining [Br<sub>2</sub>] = [Cl<sub>2</sub>] = x, the expression for [BrCl] = (1-2x). Solving for x:

$$9.1 = (1-2x)^2/x^2$$

Multiplying this gives 9.1 = (1 - 4x + 4x<sup>2</sup>)/x<sup>2</sup>; rearranging gives 5.1x<sup>2</sup> + 4x - 1 = 0.

Solving gives x = 0.2. The amounts of Br<sub>2</sub> and Cl<sub>2</sub> are both = x, so each is 0.2 (or 40%).

The amount of BrCl is (1-2x) = 0.6 (or 60%).

## B. Polarity of BrCl

This part of the discussion explores the polarity of bromine chloride and the dissociation properties that flow therefrom. Mills et al. lists a dipole moment for BrCl of 0.56 D (Table I, Page 161, right column), and indicates that the polarity of BrCl can be shown as "Br—Cl<sup>-</sup>" (Page 162, bottom of left column). This is supported by the reactivity observed for bromine chloride and shown in Scheme I of Mills et al., in which the bromine atom, not the chlorine atom, of the BrCl adds to the organic substrate for most reactions shown, except addition across a double bond, in which both atoms of the diatomic molecule are added (Page 161).

The results described in the Second Declaration of Dr. McKinnie are consistent with the observations regarding the equilibrium mixture of BrCl, Br<sub>2</sub>, and Cl<sub>2</sub> that constitutes BrCl, and their relative amounts. Paragraphs 4, 5 b-d, and 6-7 of the McKinnie Declaration are reproduced here:

4. In 2000 as part of my research duties at Albemarle I personally conducted lab

experiments to determine the N-halo compound make-up (specifically, the presence of N-bromosulfamate and N-chlorosulfamate) of a biocide solution which results from adding bromine chloride (pre-prepared or made inline) to an aqueous alkali metal sulfamate solution formed from water, sulfamic acid and sodium hydroxide (an alkali metal base). I also experimented adding bromine first, then chlorine, to the same aqueous alkali metal sulfamate solution.

5. b. N-chloro compounds are very slow to oxidize DPD but in the presence of a trace of iodide the oxidation does occur rapidly.
  - c. While N-chloro compounds are very slow to oxidize DPD in the absence of iodide, N-bromo compounds do oxidize DPD in the absence of iodide.
  - d. Thus, carrying out the analysis in the absence of iodide gives the quantity of N-bromo compound. Repeating the analysis in the presence of iodide gives the total oxidant level, which is a measure of both the N-bromo and N-chloro compounds. The ratio of the N-halo to total oxidant level represents the mole fraction of N-bromo compounds present in the final biocide solution.
6. When the pH of the aqueous alkali metal sulfamate solution prior to bromine chloride addition has a pH above 13, and upon simultaneous addition of bromine chloride with sodium hydroxide in order to maintain the pH above 13, only about 80 mole % of the N-halo sulfamate compound formed was N-bromosulfamate. The remaining about 20 mole % N-halo compound formed was N-chlorosulfamate. Similar results were achieved regardless of whether the bromine chloride was pre-prepared or made inline.
7. The above results were consistent with the fact that bromine chloride exists as an equilibrium mixture of 20 mole % Br<sub>2</sub>, 20 mole % Cl<sub>2</sub>, and 60 mole % BrCl.

The above-described experiments performed by Dr. McKinnie allowed differentiation and determination of the relative amounts of N-chlorosulfamate and N-bromosulfamate formed from BrCl in water. Further explanation of the results obtained may be in order:

- The 20% N-chlorosulfamate is from the Cl<sub>2</sub> in the equilibrium mixture.
- The Br<sub>2</sub> in the equilibrium mixture contributes 20% of the N-bromosulfamate.
- The remaining 60% N-bromosulfamate is from the BrCl (60% + 20% = 80%).

BrCl does not form N-chlorosulfamate, as discussed in the next section.

## II. Redox characteristics of BrCl

### A. Hydrolysis and BrCl

To forestall confusion in the following discussion, the reader is reminded that in a hypohalite ion or hypohalous acid (e.g., OCl<sup>-</sup>, HOBr), the halogen atom is in a formal

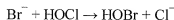
oxidation state of +1.

Literature reports agree that hypochlorite ( $\text{OCl}^-$ ) quantitatively oxidizes bromide ( $\text{Br}^-$ ) to hypobromite ( $\text{OBr}^-$ ):

Mills et al. discusses the reaction of bromine chloride with water, stating that "Bromine chloride appears to hydrolyze exclusively to hypobromous acid",  $\text{BrCl} + \text{H}_2\text{O} \rightarrow \text{HOBr} + \text{HCl}$  (Page 164, right column). Mills et al. further states that "If any hydrobromic acid (HBr) were formed by hydrolysis of the dissociated bromine, it would be quickly oxidized by hypochlorous acid to hypobromous acid,  $\text{HBr} + \text{HOCl} \rightarrow \text{HOBr} + \text{HCl}$ .

Farkas and Lewin observe that "upon the addition of bromide, all the hypochlorite was converted to hypobromite" (Page 5766, right column, second paragraph). In chemical words,  $\text{OCl}^- + \text{Br}^- \rightarrow \text{OBr}^- + \text{Cl}^-$ . The reaction is apparently irreversible. Farkas and Lewin monitored the reaction via the change in pH, stating that "The final pH was established within a few seconds after the addition of the bromide and remained constant for more than 15 minutes" (Page 5766, right column, third paragraph).

Harp, at Page 12, right column, lines 18-22, states that "The addition of chlorine [ $\text{Cl}_2$ ] to water containing bromide [ $\text{Br}^-$ ] will produce hypobromous acid and hypobromite ion:



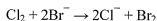
*This reaction is irreversible.*" (Emphasis added.) For completeness, it is mentioned that chlorine hydrolyzes in water:  $\text{Cl}_2 + \text{H}_2\text{O} \rightarrow \text{HOCl} + \text{HCl}$ .

#### **B. Non-formation of $\text{BrCl}$ (from $\text{Cl}^-$ and $\text{OBr}^-$ )**

Another way of phrasing the title of this section is  $\text{Cl}^- + \text{Br}^+ \nrightarrow \text{Cl}^+ + \text{Br}^-$ . This issue has previously been discussed, at least in part, on Pages 6-7 of the Response filed on August 4, 2009, where the electrochemical infeasibility of oxidation of  $\text{Br}^-$  by  $\text{Cl}^+$  (in the form of  $\text{HOCl}$ ) was discussed. That discussion is incorporated by reference herein.

The web page entitled "Halogens as Oxidizing Agents" is instructive. First, it explains that

In each case, a halogen higher in the group can oxidise the ions of one lower down. For example, chlorine can oxidise the bromide ions (in, for example, potassium bromide solution) to bromine:



(printed Page 2, first two sentences; first equation shown.)

This web page further explains that

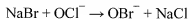
Bromine can only oxidise iodide ions to iodine. **It [bromine] isn't a strong enough oxidising agent to convert chloride ions into chlorine.** (printed Page 2, paragraph under Note; emphasis added.)

The remainder of the web page describes the reasons for the differing oxidative abilities of the various halogens.

### III. Application to the Rejection of the Present Claims

As pointed out in previous Responses, the aqueous biocidal compositions of the present claims comprise N-bromosulfamate and N-chlorosulfamate since they are made from BrCl, while the compositions produced by Goodenough comprise only N-bromosulfamate, since they are made from Br<sub>2</sub>. All of the above discussion supports and clarifies that BrCl is not produced inherently or otherwise in the solutions of Goodenough.

Dallmier relies on the oxidation of bromide to hypobromite by hypochlorite:



If this reaction were reversible, the process of Dallmier, in which waiting for a period of time is expressly included as one of the steps of the process, would not work very well. The goal in Dallmier is to make a bromine-based biocide. Thus Dallmier, cited in the rejection of the present claims, lends support to the absence oxidized chlorine species and therefore the absence of N-chlorosulfamate in Goodenough.

One further aspect of the present claims that has not been addressed in this response is the pH of the solutions. Goodenough teaches a pH of 8-10, and indicates that

the hydroxide used to attain the desired pH, e.g., NaOH or  $\text{Mg}(\text{OH})_2$ , is not important. This has been addressed in the First Declaration of Dr. McKinnie and in the Nalepa Declaration, as follows:

In the First Declaration of Dr. McKinnie, the details of Goodenough's Example 3 are described in paragraphs 101-103 of the First Declaration. (*Note: Exhibit 1003 is the Goodenough patent; references to column and line therein are to column and line in Goodenough*). As noted in paragraph 101, the hydroxide used in Example 3 of Goodenough was magnesium hydroxide, an alkaline earth metal hydroxide. *The low solubility of alkaline earth metal hydroxides in water makes it impossible to produce an aqueous solution having a pH greater than or equal to 13 when alkaline earth metal hydroxide is the only hydroxide source.* (First Declaration, paragraph 47b.) Thus, Example 3 of Goodenough does not describe a method utilizing an initial pH about 13.0 or greater, and certainly does not describe a method wherein a pH of about 13.0 or greater is maintained during addition of bromine. (Emphasis added.)

The Nalepa Declaration reports on a repeat of the preparation of Solution B from Goodenough's Example 3, both as described in Goodenough with  $\text{Mg}(\text{OH})_2$ , and by replacing the  $\text{Mg}(\text{OH})_2$  with an equimolar amount (based on hydroxide) of NaOH. The pH of the solution prepared with  $\text{Mg}(\text{OH})_2$  was 8.75, while the solution prepared with NaOH had a pH of 11.55. *In these experiments, a final pH of 13 or greater was not attained with either hydroxide.* Thus, Goodenough does not show solutions having a pH of about 13 or greater, regardless of whether the hydroxide is  $\text{Mg}(\text{OH})_2$  or an alkali metal hydroxide (paragraph 6, Table 1; emphasis added).

Recalling that the present claims stand rejected for obviousness, the Nalepa Declaration also demonstrates that unexpected results have been obtained:

For the biocides of Goodenough, the stability is markedly lower for solution made with sodium hydroxide. After four days, the solution prepared with NaOH retained only 69% of its original activity, while the solution prepared with  $\text{Mg}(\text{OH})_2$  as in Goodenough retained 97% of its original activity (paragraph 9, Table 2, reproduced below).

Procedure	Solution	Initial activity	Activity after 4 days	Activity retained
Goodenough	Solution 1 - $\text{Mg}(\text{OH})_2$	9560 ppm $\text{Br}_2$	9290 ppm $\text{Br}_2$	97%
Goodenough	Solution 2 - $\text{NaOH}$	9610 ppm $\text{Br}_2$	6600 ppm $\text{Br}_2$	69%
Present invention	Solution 3 - $\text{NaOH}$	149,000 ppm $\text{Br}_2$	150,000 ppm $\text{Br}_2$	99%

Thus, the teachings of Goodenough indicate that alkali metal bases provide solutions which are inferior for retention of activity. This indicates that the presently claimed solutions do provide unexpected results – significantly higher retention of activity (99% after 4 days) while using an alkali metal base (Nalepa Declaration, Table 2).

#### **Acknowledgement of previously submitted IDS**

Applicants note with appreciation the indication that the Examiner has considered the Information Disclosure Statement submitted on May 9, 2003.

In light of the foregoing remarks, the case is believed to be in condition for allowance. Prompt notification to this effect would be sincerely appreciated.

If any matters remain that require further consideration, the Examiner is requested to telephone the undersigned at the number given below so that such matters may be discussed, and if possible, promptly resolved.



Please continue to address all correspondence in this Application to Albemarle Corporation, at the address of record.

Respectfully submitted,

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